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Microwave shielding of ultracold polar molecules with imperfectly circular polarization

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We investigate the use of microwave radiation to produce a repulsive shield between pairs of ultracold polar molecules and prevent collisional losses that occur when molecular pairs reach short range. We carry out coupled-channels calculations on RbCs + RbCs and CaF + CaF collisions in microwave fields. We show that effective shielding requires predominantly circular polarization but can still be achieved with elliptical polarization that is around 90% circular.

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I. INTRODUCTION

Ultracold polar molecules provide many new opportunities for quantum science, and may in future provide a new platform for quantum technology. Several groups have succeeded in producing samples of such molecules, either by direct laser cooling of preexisting molecules [1,2] or by associating pairs of ultracold atoms and transferring the resulting weakly bound molecules to the ground state [3–10].

A few groups have carried out experiments on collisions of ultracold polar molecules. Fermionic molecules in identical states are protected against collisions at the lowest temperatures [9,11], but all bosonic species investigated so far have been found to undergo fast collisional trap loss. The same is true for fermionic molecules in different internal states. For some molecules, such as KRb [11], the loss may be attributed to energetically allowed two-body reactions. However, fast loss with second-order kinetics appears to exist [12,13] even when there are no energetically allowed two-body pathways [14]. In such cases the loss may be mediated by formation of long-lived collision complexes, followed by either collision with a third molecule [15] or excitation by the trapping laser [16,17].

It thus appears that active measures are needed to stabilize gases of ultracold polar molecules against collisions. An appealing possibility is microwave shielding with blue-detuned microwave radiation [18,19], which provides a repulsive shield that prevents the molecules coming close together. We have recently shown that such shielding can be achieved with circularly polarized microwaves at high Rabi frequencies, but not with linearly polarized microwaves [19].

Pure circular polarization is hard to achieve for microwaves at the frequencies and intensities needed for shielding. In this paper we explore the effect of imperfectly circular polarization and show that good shielding can be achieved with elliptical polarization that is around 90% circular.

II. THEORY

The calculations carried out in the present paper use an extension of the methods described in Ref. [19]. We consider a collision between two polar molecules, each dressed with microwave radiation that is blue detuned by Δ from its $n = 0 \rightarrow 1$ rotational transition. The intensity of the microwave radiation is specified by the Rabi frequency Ω . The role of electron and nuclear spins is discussed below.

Our basic physical picture is that two molecules interact at long range via the dipole-dipole interaction. In the presence of blue-detuned microwave radiation, this provides a repulsive shield. Under conditions where shielding is effective, most molecules are reflected at long range, and do not approach each other close enough for short-range processes to occur. There are nevertheless two processes that can cause loss. First, any colliding pairs that do reach short range are likely to be lost. We characterize this by the probability of reaching short range (RSR), which may be expressed as a rate coefficient only in the limit of 100% loss at short range. Secondly, even pairs that are reflected at long range may undergo inelastic transitions to lower-lying field-dressed states. These transfer internal energy into relative kinetic energy and generally eject both collision partners from the trap. We refer to this latter process as microwave-induced loss.

To model shielding and loss, we carry out coupled-channels scattering calculations. We propagate two sets of linearly independent solutions of the coupled-channels equations, using the renormalized Numerov method [20], and apply both capture boundary conditions at short range and S -matrix boundary conditions at long range [21–23]. We calculate both the probability of RSR and the rate coefficient for microwave-induced loss. The remaining flux is shielded and scatters elastically.

The molecules are modeled as rigid rotors with a dipole moment. The monomer Hamiltonian of molecule X is

$$\hat{H}^{(X)} = b_{\text{rot}} \hat{n}^2 + \hat{H}_{\text{hyperfine}}^{(X)} + \hat{H}_{\text{ac}}^{(X)}. \quad (1)$$

The first term describes the rotational kinetic energy, with rotational constant b_{rot} . The second term describes the molecular

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hyperfine Hamiltonian, and is discussed in the Supplemental Material of Ref. [19]. The last term represents the interaction with a microwave electric field [24]:

$$\hat{H}_{\text{ac}}^{(X)} = -\sqrt{\frac{\hbar\omega}{2\epsilon_0 V_0}} [\hat{\mu}_\sigma^{(X)} \hat{a}_\sigma + \hat{\mu}_\sigma^{(X)\dagger} \hat{a}_\sigma^\dagger] + \hbar\omega(\hat{a}_\sigma^\dagger \hat{a}_\sigma - N_0). \quad (2)$$

Here, $N_0 = \epsilon_0 E_{\text{ac}}^2 V_0 / 2\hbar\omega$ is the reference number of photons in a reference volume, V_0 , at microwave electric field strength, E_{ac} [25]. The operators \hat{a}_σ^\dagger and \hat{a}_σ are creation and annihilation operators for photons in polarization mode σ and angular frequency ω . The polarization may be linear, with Cartesian components $\sigma = x, y$, and/or z ; circular, with spherical components $\sigma = \pm 1$ and $\hat{\mu}_{\pm 1}^{(X)} = \mp(\hat{\mu}_x^{(X)} \pm i\hat{\mu}_y^{(X)})/\sqrt{2}$; or elliptical. A general elliptical polarization in the xy plane can be described as

$$\sigma(\xi) = \sigma_+ \cos \xi - \sigma_- \sin \xi. \quad (3)$$

The ellipticity angle ξ interpolates between pure right-hand circular polarization, σ_+ , at $\xi = 0$ and linear x polarization, σ_x , at $\xi = \pi/4$. The form of the interaction with a microwave electric field, Eq. (2), remains valid with the substitution $\hat{\mu}_\sigma \rightarrow \hat{\mu}_{+1} \cos \xi - \hat{\mu}_{-1} \sin \xi$.

The total Hamiltonian is

$$\hat{H} = -\frac{\hbar^2}{2M} \frac{1}{R} \frac{d^2}{dR^2} R + \frac{\hbar^2 \hat{L}^2}{2MR^2} + \hat{H}^{(A)} + \hat{H}^{(B)} + \hat{V}(R). \quad (4)$$

Here M is the reduced mass, R is the distance between the molecules, and \hat{L} is the dimensionless angular momentum operator associated with the end-over-end rotation of the intermolecular axis, \vec{R} . The first term describes the radial kinetic energy and the second term describes the centrifugal kinetic energy. The third and fourth terms correspond to the monomer Hamiltonian of Eq. (2). The final term is the interaction potential $\hat{V}(R)$, which in the present paper is limited to the dipole-dipole interaction [19],

$$\hat{V}(R) = -\frac{\sqrt{6}}{4\pi\epsilon_0 R^3} T^{(2)}(\hat{\mu}^{(A)}, \hat{\mu}^{(B)}) \cdot C^{(2)}(\hat{R}), \quad (5)$$

where $C^{(2)}(\hat{R})$ and $T^{(2)}(\hat{\mu}^{(A)}, \hat{\mu}^{(B)})$ are second-rank spherical tensors the normalizations and scalar product of which are given in the Supplemental Material of Ref. [19].

We use completely uncoupled basis sets. The basis set for monomer $X = A, B$ consists of products of rotational, spin, and photon states,

$$|n_X m_{i_X}\rangle |i_X m_{i_X}\rangle |N\rangle, \quad (6)$$

where $|i_X m_{i_X}\rangle$ schematically represents all the spins on molecule X ; it may be a product of spin functions, including electron spin where necessary. N is the number of photons with respect to N_0 . The blue detuning is given by positive Δ . For the colliding pair of molecules we introduce an angular momentum state, $|LM_L\rangle$, that describes the end-over-end rotation of the intermolecular axis. The basis functions for the pair of molecules are [26–28]

$$|n_A m_{i_A}\rangle |i_A m_{i_A}\rangle |n_B m_{i_B}\rangle |i_B m_{i_B}\rangle |LM_L\rangle |N\rangle. \quad (7)$$

Only even values of L are included: The only interaction that couples states with different L is the dipole-dipole interaction,

and this conserves the parity of L . The basis functions are adapted to permutation symmetry as described in the Supplemental Material of Ref. [19].

Excluding the photons, the direct-product basis functions have a well-defined projection quantum number M_{tot} for the projection of the total angular momentum along the space-fixed z axis:

$$M_{\text{tot}} = m_{n_A} + m_{i_A} + m_{n_B} + m_{i_B} + M_L. \quad (8)$$

For linear z or circular $x \pm iy$ polarizations, with $\sigma = 0, \pm 1$, the generalized projection quantum number \mathcal{M} is conserved,

$$\mathcal{M} = M_{\text{tot}} + \sigma N, \quad (9)$$

and the basis set can be limited to include only functions with a single value of \mathcal{M} . For elliptical polarization in the xy plane, however, there is no generalized projection quantum number that is conserved. In this case, the basis set must include functions with various values of \mathcal{M} , and the calculation is correspondingly more computer intensive.

We showed in Ref. [19] that molecular fine and hyperfine interactions cause an increase in microwave-induced loss at zero magnetic field, but that the increase can be suppressed by applying a moderate magnetic field perpendicular to the plane of polarization. We demonstrate in the Supplemental Material [29] that the field required to suppress the effects of hyperfine interactions is proportional to the ratio of the hyperfine constants to the nuclear g factors. For molecules in $^2\Sigma$ states, the field needed is proportional to the ratio of the spin-rotation constant to the electron g factor. Since there is a tendency for both hyperfine coupling constants and nuclear g factors to increase with atomic number, the magnetic field required to suppress hyperfine effects does not vary enormously between systems, and is typically on the order of 100 G. The key issue is that the magnetic field must be high enough that m_n is nearly conserved. When it is, the microwave-induced loss for the alkali-metal diatomic molecules is reduced to the hyperfine-free level, while that for molecules in $^2\Sigma$ states is incompletely suppressed [29].

III. RESULTS

We have carried out coupled-channel calculations for RbCs + RbCs and CaF + CaF collisions in microwave fields, with the molecules in their ground electronic states, $X^1\Sigma^+$ and $X^2\Sigma^+$, respectively. The calculations used $n_{\text{max}} = 1$; $L_{\text{max}} = 6$; $N = 0, -1, -2$; and a collision energy of 1 μK . Electron and nuclear spins were not included explicitly, but for CaF the calculation included spin-dependent couplings averaged over the spin-stretched state. As shown in the Supplemental Material, these calculations are appropriate for magnetic fields above 100 G [29].

Figure 1 shows loss processes for RbCs as a function of the ellipticity angle, ξ , and the Rabi frequency, Ω , for fixed $\Delta = 0$. Panel (a) shows the probability of RSR, and panel (b) shows the rate coefficient for microwave-induced loss, i.e., inelastic transitions to lower-lying field-dressed states. Figure 1 shows the probability of RSR and the rate coefficient for microwave-induced loss for RbCs as a function of the ellipticity angle, ξ , and the Rabi frequency, Ω , for fixed $\Delta = 0$.

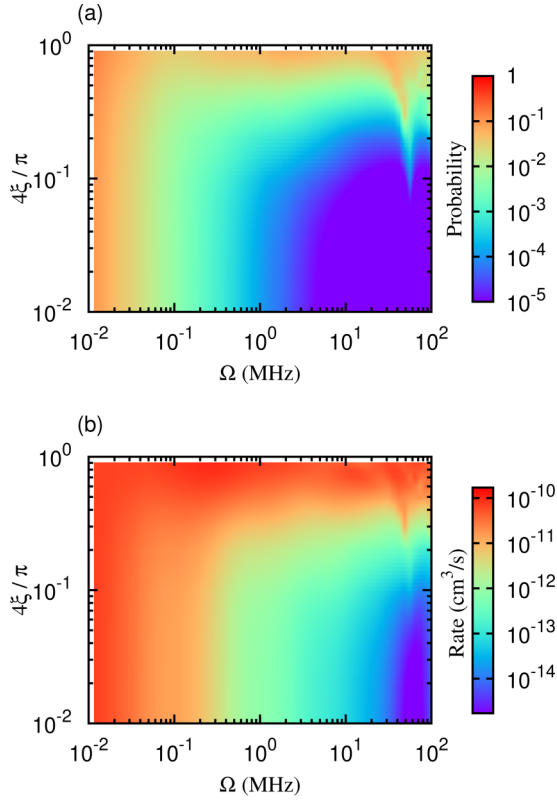


FIG. 1. Probability of RSR (a) and rate coefficient for microwave-induced loss (b) in RbCs + RbCs collisions with microwave detuning $\Delta = 0$, as a function of Rabi frequency, Ω , and ellipticity angle, ξ .

At the top of the figures, where $4\xi/\pi \approx 1$, the probability of RSR and the microwave-induced loss rate are both large. This corresponds to linear polarization, for which shielding is ineffective. At the bottom of the figure, below $4\xi/\pi \approx 0.01$, the probability of RSR and the microwave-induced loss rate become independent of ξ and visually indistinguishable from the result for circular polarization. Losses can be suppressed to four orders of magnitude below the universal loss rate [30], which for RbCs at zero energy is $1.7 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$. These values of ξ may be experimentally realizable. An ellipticity of $4\xi/\pi \approx 0.1$, corresponding to a microwave field that is 90% circular and a power ratio around 20 dB between σ_+ and σ_- polarizations, results in somewhat increased losses but still provides effective shielding.

Figure 2 shows the probability of RSR and the rate coefficient for the microwave-induced rate for CaF as a function of the ellipticity angle, ξ , and the Rabi frequency, Ω , for fixed $\Delta = 0$. The spin degrees of freedom are not explicitly accounted for, but the calculation includes fine and hyperfine couplings averaged over the spin-stretched state, which is appropriate for magnetic fields above 100 G [29]. The resulting couplings increase the microwave-induced loss. The resulting shielding is less effective than for RbCs, but the losses are still reduced by up to three orders of magnitude from the universal loss rate of $5 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$. The loss rates are essentially indistinguishable from those for circular polarization for $4\xi/\pi \leq 0.1$.

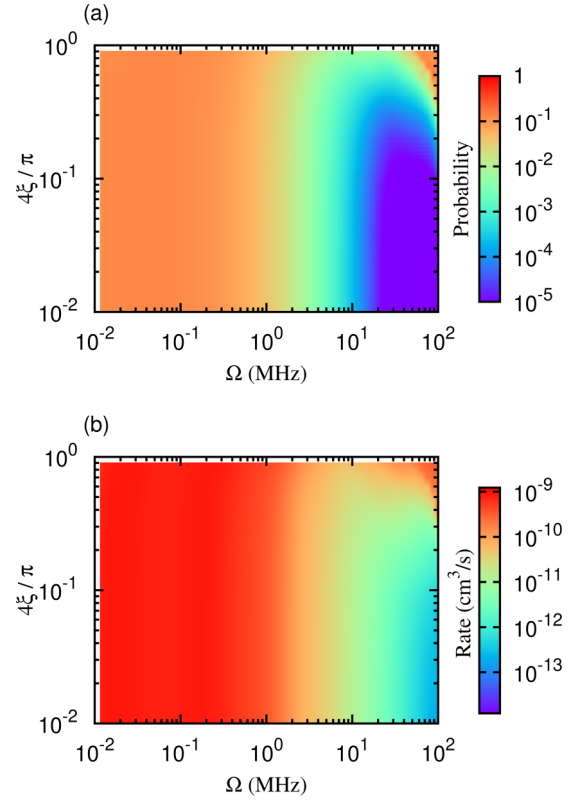


FIG. 2. Probability of RSR (a) and rate coefficient for microwave-induced loss (b) in CaF + CaF collisions with microwave detuning $\Delta = 0$, as a function of Rabi frequency, Ω , and ellipticity angle, ξ .

IV. FIXED- θ COLLISIONS (SUDDEN APPROXIMATION)

To understand the way that the losses depend on the ellipticity angle, we perform additional calculations using a sudden approximation in which the orientation of the intermolecular axis is assumed to remain constant during a collision. The orientation is specified by the polar angles (θ, ϕ) of \hat{R} with respect to the space-fixed frame. The spherical harmonics $|L, M_L\rangle$ are dropped from the basis set. This approximation is expected to be accurate if the period of an end-over-end rotation of the complex is long compared to the duration of a collision, which may not be the case. It also neglects nonadiabatic losses due to couplings involving $d/d\theta$, which may be significant. The fixed- θ approximation is therefore not quantitatively accurate, but it nevertheless provides useful insights. The results presented in Sec. III were obtained from full coupled-channels calculations that do not use the sudden approximation.

Figure 3 shows adiabatic potential curves for RbCs + RbCs, defined as eigenvalues of the pair Hamiltonian, Eq. (4), excluding radial kinetic energy, as a function of R for fixed θ . This representation is much simpler than is obtained by diagonalizing in the full basis set that includes the dependence on θ through the partial-wave expansion, as shown in Fig. 3 of the Supplemental Material of Ref. [19]. Figure 3 provides a less crowded representation of the adiabats that removes many of the inconsequential crossings of low- and high- L states corresponding to different thresholds. In each case

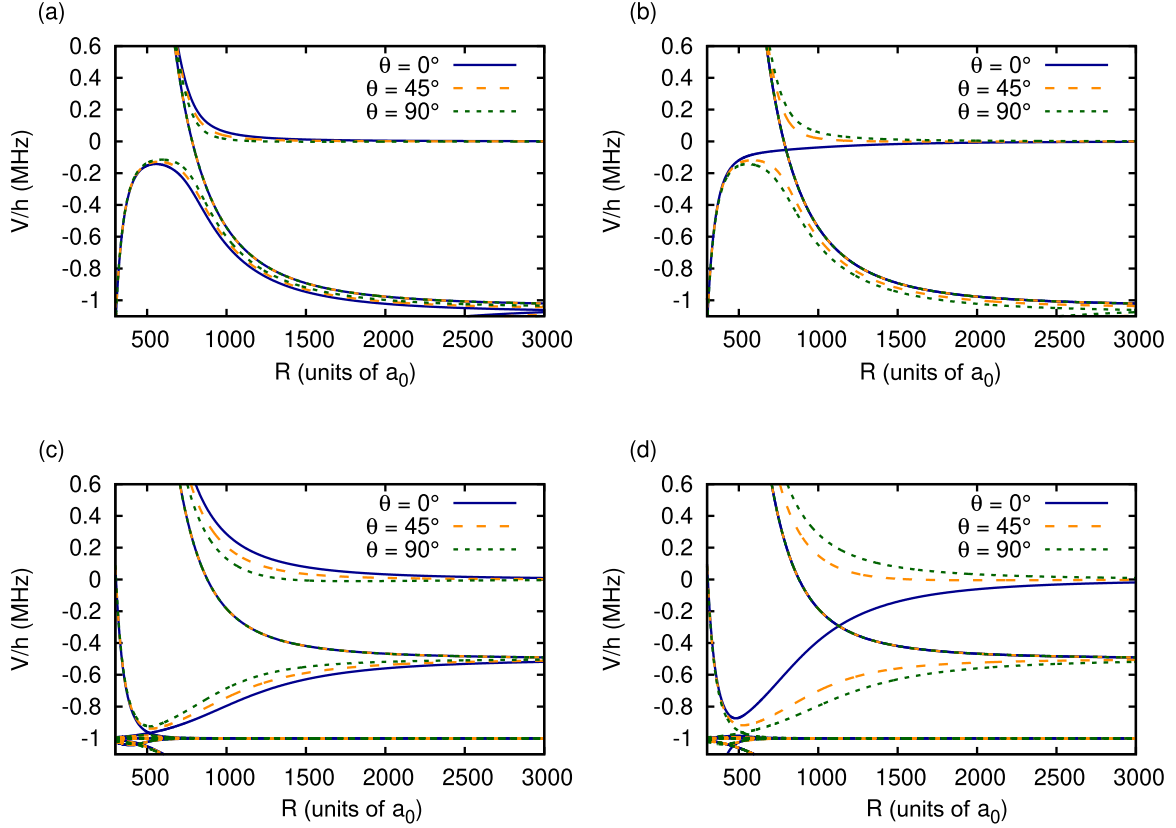


FIG. 3. Adiabatic potential curves for RbCs + RbCs with $\Omega = 0.2$ MHz and fixed orientation θ as indicated in the legend. The different panels correspond to (a) circular polarization for $\Omega \ll \Delta$, (b) linear polarization for $\Omega \ll \Delta$, (c) circular polarization for $\Omega \gg \Delta$, and (d) linear polarization for $\Omega \gg \Delta$.

the colliding molecules are initially at the uppermost of the thresholds shown.

The upper two panels of Fig. 3 show adiabats for the off-resonant case, $\Omega \ll \Delta$. This is not the parameter regime in which effective shielding is realized, but the potential curves are more easily understood. In this case the ground-state ($|n_A, n_B, N\rangle = |0, 0, 0\rangle$) and microwave-dressed excited-state ($|0, 1, -1\rangle$) potentials are essentially unmodified by the microwaves. The ground-state (upper) potential is determined by rotational dispersion and varies with R^{-6} . The excited-state (lower) potential is split by resonant dipole-dipole interactions into an attractive and repulsive branch, which vary with R^{-3} ; the attractive branch is mostly off scale in Fig. 3. Resonant dipole-dipole interactions quantize the rotational angular momentum along the intermolecular axis with projection quantum number K , giving potentials that are independent of orientation with respect to the microwave polarization: $K = 0$ for the attractive branch and $K = \pm 1$ for the repulsive branch. In this off-resonance scenario, coupling by the microwaves is significant only near the point where the ground-state potential crosses the repulsive $K = \pm 1$ branch of the excited state, around $800 a_0$ in Fig. 3. For linear π or circular σ_{\pm} polarization, the microwave field couples the ground state to the excited state with well-defined space-fixed projection quantum number $M_N = m_{nA} + m_{nB} = 0$ or ± 1 , respectively. Hence, the coupling to the repulsive $K = \pm 1$ branch depends on the polar angle of the intermolecular axis, θ , as the Wigner d function $d_{M_N, K}^{(1)}(\theta)$. For circular σ_{\pm} polarization, shown in

Fig. 3(a), the crossing is avoided for all angles θ , although the degree of avoidedness is anisotropic and varies with $\sqrt{1 + \cos^2 \theta}$. For linear π polarization, shown in Fig. 3(b), the avoidedness of the crossing varies with $\sin \theta$, and vanishes at $\theta = 0$. This implies that there is a “hole” in the shielding at $\theta = 0$ with linear polarization.

Next, we consider the resonant case, $\Omega \gg \Delta = 0$, where effective shielding is achieved with near-circular polarization. The adiabats are shown in the lower two panels of Fig. 3. Here, there is strong dressing that mixes the $n = 0$ ground and $n = 1$ excited states even for the isolated molecules. This can be interpreted as inducing an oscillating dipole moment in the laboratory frame, which produces first-order dipole-dipole interactions upon time averaging. As a result, the adiabats vary with $R^{-3} P_2(\cos \theta)$ at long range. The space-fixed dipole-dipole interactions are attractive for some orientations, i.e., near $\theta = 0$ for linear π polarization and near $\theta = 90^\circ$ for circular σ_{\pm} polarization. However, at shorter separation, the dipole-dipole interaction becomes stronger than $\hbar\Omega$, and this interaction again quantizes the molecules along the intermolecular axis, such that the top adiabatic curve is repulsive for all θ . As discussed above, the coupling from the initial state to this repulsive branch depends on θ . For circular polarization, shown in Fig. 3(c), the angle dependence is again such that the coupling to the repulsive branch never vanishes. For linear polarization, shown in Fig. 3(d), the shielding again vanishes at $\theta = 0$, where both $K = \pm 1$ repulsive states are uncoupled from the initial state. For small θ , this leads to a

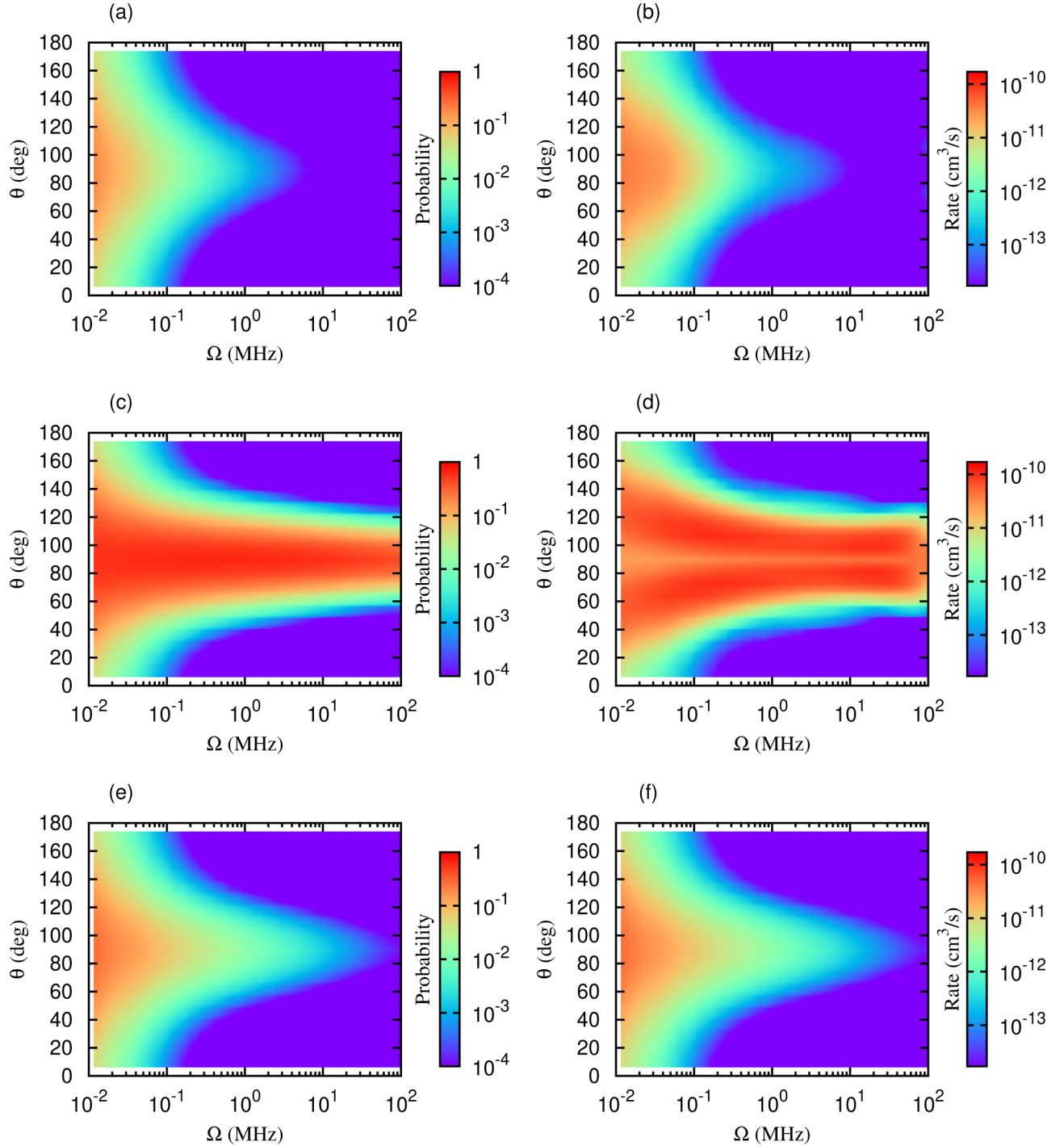


FIG. 4. Probabilities for reaching short range [(a), (c), (e), left-hand column] and rates of microwave-induced loss [(b), (d), (f), right-hand column] as a function of Rabi frequency and fixed orientation θ . Panels (a) and (b) are for σ^+ circular polarization, panels (c) and (d) are for σ_x linear polarization, and panels (e) and (f) are for elliptical polarization with $\xi = 0.2\pi/4$.

narrowly avoided crossing between the initial state and the lower field-dressed level; this occurs outside the repulsive shield and is classically accessible. As a result, linear polarization does not achieve shielding, but it does give rise to additional microwave-induced losses. For imperfectly circular polarization, the effective coupling to the repulsive branch is reduced from that for circular polarization, but does not vanish.

It is worth noting that the space-fixed dipole-dipole interaction discussed here produces attractive interactions outside the repulsive shield. Where these support bound states, they can be used to tune the scattering length while shielding from short-range losses, as shown in Ref. [31].

We next carry out coupled-channels calculation of shielding and loss in the fixed- θ approximation. For these we consider elliptical polarization in the xy plane, with $\xi = \pi/4$

corresponding to linear polarization along x (σ_x polarization). This differs from the discussion above, which considered linear polarization along z (π polarization). Since only the microwave polarization defines a preferential direction, this choice is inconsequential, but it does mean that collisions now occur along the axis of linear polarization when $\theta = 90^\circ$ rather than $\theta = 0$.

Figure 4 shows the probabilities of RSR and the rate coefficients for microwave-induced loss in RbCs + RbCs collisions, for $\Delta = 0$, as a function of the Rabi frequency, Ω , and the fixed angle, θ , between the intermolecular axis and microwave propagation direction. The three rows correspond to different polarizations: σ_+ , σ_x , and elliptical polarization with $\xi = 0.2\pi/4$. For circular polarization, shown in the top row, we find both types of loss can be suppressed by using sufficiently high Rabi frequency. The loss is highest for $\theta = 90^\circ$, where coupling to the repulsive branch of the resonant dipole-dipole interaction is weakest and long-range space-fixed dipole-dipole interactions are attractive. For linear x polarization, shown in the center row, we find losses are also highest near $\theta = 90^\circ$, where unshielded collisions along the polarization direction occur, as discussed above. These losses cannot be reduced by increasing the Rabi frequency. The bottom row shows results for elliptical polarization with $\xi = 0.2\pi/4$. The results are qualitatively similar to those in the case of circular polarization, but somewhat higher Rabi frequencies are required to suppress the loss.

V. CONCLUSIONS

We have investigated shielding of ultracold molecular collisions using microwave radiation with imperfectly circular polarization. The goal of microwave shielding is to prevent colliding pairs of molecules from reaching short range, where losses occur with high probability, while simultaneously avoiding long-range losses induced by the microwaves themselves. We have carried out coupled-channels calculations on RbCs + RbCs and CaF + CaF collisions to evaluate both the probabilities of reaching short range and the rate coefficients for microwave-induced loss.

We showed previously [19] that effective shielding can be achieved with circularly polarized microwaves but not

with linearly polarized microwaves. However, pure circular polarization is hard to achieve in an apparatus designed for trapping ultracold molecules. Here we investigate how the effectiveness of shielding degrades for imperfectly circular polarization. We show that effective shielding can still be achieved with elliptical polarization corresponding to a microwave field that is around 90% circular, with a power ratio around 20 dB between σ_+ and σ_- polarizations.

Molecular fine and hyperfine structure can interfere with microwave shielding. We show that, for molecules in $^1\Sigma$ states, such as the alkali-metal diatomic molecules, effective shielding can be restored by applying a moderate magnetic field, of order 100 G, perpendicular to the plane of polarization. For molecules in $^2\Sigma$ states, such as CaF, shielding is not fully restored by a magnetic field but losses can still be reduced by up to three orders of magnitude compared to the universal limit of complete short-range loss.

We interpret our results in terms of an approximate model in which collisions occur at a fixed orientation of the intermolecular axis. This simplifies the effective potential curves that govern shielding and loss. For linear polarization, there are two curves in this model that cross (without an avoided crossing) when a collision occurs with the intermolecular axis along the axis of polarization. This is responsible for the lack of shielding for linear polarization. Elliptical polarization turns this crossing into an avoided crossing, which increases in strength as the degree of circular polarization increases. This restores effective shielding even for imperfectly circular polarization.

We anticipate that microwave shielding with near-circular polarization will be a valuable tool for reducing collisional losses in samples of ultracold polar molecules.

Data presented in this paper are available at Ref. [32].

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